

# Argonne National Laboratory

THE ATTACK ON THE CLADDING OF  
EBR-II DRIVER FUEL ELEMENT C-249-16

by

W. E. Ruther, T. D. Claar,  
and S. Matras

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EBR-II Project

March 1969





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# THE ATTACK OF THE CLADDING OF EBR-II DRIVER FUEL ELEMENT C-249-16

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## ABSTRACT

An investigation was made to determine the cause of grain-boundary attack in selective areas of the Type 304L stainless steel cladding of a driver fuel element removed from EBR-II. A hypothesis that improper cleaning techniques may have caused the damage was rejected after variations of normal cleaning methods failed to duplicate the phenomenon. Chemical and electron-microprobe analyses, along with metallographic work, indicated that an increase in carbon concentration was associated with the areas of selective attack. Further testing showed that the microstructure of a stainless steel surface that had been intentionally contaminated with a carbonaceous substance before exposure to sodium was essentially the same as the microstructure of the cladding that had been exposed in EBR-II.

## I. INTRODUCTION

Areas of deep selective grain-boundary attack were found in the Type 304L stainless steel cladding of one spent Mark-IA driver element during a metallographic study of fuel/cladding compatibility. This element was one of four Mark-IA elements that had been selected for examination to detect possible fuel/cladding interactions because they showed relatively high fuel swelling, ranging from 10.9 to 14.8% (as indicated by postirradiation measurements of sodium level). Metallographic sections of the cladding of element 16 from subassembly C-249 (identified as element C-249-16 hereafter) were found to have several axial grain-boundary fissures originating in the outer surface of the cladding and penetrating into the cladding. Most of the fissures were shallow (see Fig. 1),\* but the appearance of one metallographic section can be interpreted to indicate penetration through the cladding (see Fig. 2). The fissures were noted only in that part of the element where maximum fuel swelling occurred.

\*Unless indicated otherwise, all specimens were etched for 15 sec in 5% oxalic acid, and all photographs were taken at a magnification of 400X and reduced photographically by 10%.



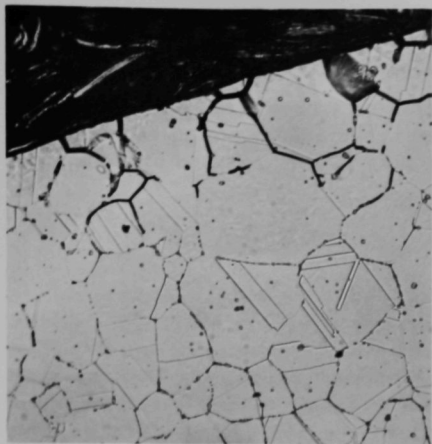


Fig. 1. Cladding of Element C-249-16 after 45-sec Etch in 5% Oxalic Acid

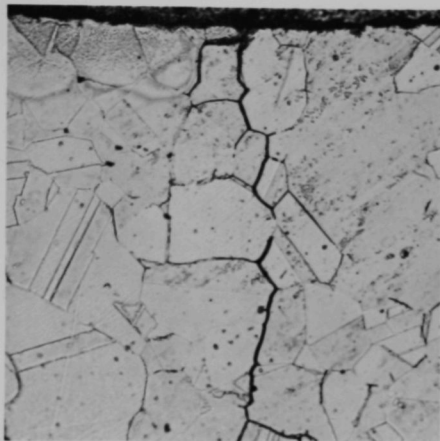


Fig. 2. Cladding Crack in Specimen 46A2 from Element C-249-16. Etched in  $\text{HNO}_3\text{-HCl-glycerol}$ .

The cladding came from a lot of seamless, air-melted material, most of which had been used in about 3500 fuel elements installed in the reactor by the end of 1966. Element C-249-16 was in the reactor from July 16, 1966 to December 4, 1966. There is no evidence that this lot of cladding material influenced reactor performance, because (a) no fission-product release was detected, (b) no core or subassembly malperformance occurred, and (c) no loss of bond sodium or other visible effects were observed in the spent fuel.

The spent element was stored outside the reactor for about eight months before it was shipped to Illinois for examination. The possibility that the storage environment or cleaning procedure contributed to the intergranular fissures was considered.

Previous metallographic investigations and regular EBR-II surveillance studies were rechecked for indications of similar attack. None was found. Sixteen additional elements (over 80 polished sections) were examined, but none of them showed indications of such attack. Furthermore, tube-burst tests of the cladding from 20 fuel elements of seven subassemblies removed from the reactor in 1965 and early 1966 had shown no evidence of cracks. Although the preponderance of evidence indicated that the attack of the C-249-16 cladding was unique, the potential hazard to the reactor required that a careful investigation be made to determine the most probable cause of this near-failure. A task group was organized under the direction of R. A. Noland. Through the cooperation of members of this group, two hypotheses of the cause of the attack were postulated and evaluated:



1. Slight variations in the normal cleaning or storage of the fuel element could have exposed heat-sensitized stainless steel to concentrated sodium hydroxide solution for a period of time.

2. The grain boundaries could have been selectively carburized before or during the exposure to reactor sodium, making the boundaries susceptible to cracking and etchant attack.

## II. EXPERIMENTAL PROCEDURE

Seamless Type 304L stainless steel tubing from the lot used to fabricate the cladding of element C-249-16 was exposed to sodium in the ex-reactor recirculating system shown in Fig. 3. (Table I gives the chemical analysis and mechanical properties of the stainless steel used in the studies.) For comparison, welded tubing from a similar lot was exposed in the same system. Standard vibratory polishing techniques were used in the metallographic preparation of specimens,\* which were subsequently etched electrolytically with a 5-wt % solution of oxalic acid.

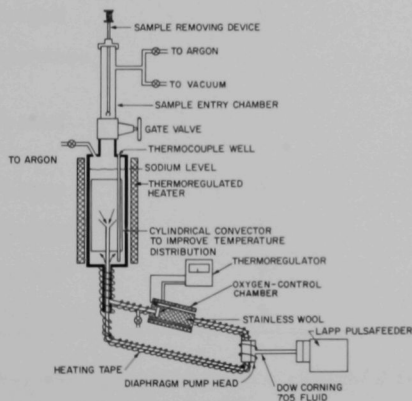


Fig. 3

Recirculating System in  
Which Specimens Were  
Exposed to Sodium

TABLE I. Type 304L Stainless Steel Used in  
Carburization Studies

Chemical Analysis, wt %		Mechanical Properties	
C: 0.02	Si: 0.40	Yield strength:	39,000 psi
Mn: 1.0	Cr: 18.8	Ultimate strength:	90,000 psi
P: 0.006	Ni: 10.0	Elongation:	55% in 2 in.
S: 0.010		Hardness:	Rockwell B-69

\*S. Matras, Vibratory Grinding and Polishing of Metallographic Specimens, ANL-6975 (Nov 1965).



### A. Cleaning Hypothesis

Several tests were conducted on specimens exposed to 550°C sodium and then cleaned by different techniques.

Table II lists the test specimens and the techniques used to clean them. For each group listed, five samples were prepared: (1) electro-polished; (2) as-received, transverse; (3) as-received, longitudinal; (4) plastically deformed, transverse, and (5) heat-sensitized. The stainless steel specimens were sensitized by exposing them to a temperature of 650°C for 4 hr under vacuum.

TABLE II. Ex-reactor Sodium Exposures and Cleaning Procedures

Group	Tube	Duration of Exposure to 550°C Sodium, weeks	Cleaning Treatment before Microexamination
1	Seamless	1	Moist argon
2	Welded	1	Moist argon
3	Seamless	1	Moist argon; 1 week storage in laboratory atmosphere
4	Welded	1	Moist argon; 1 week storage in laboratory atmosphere
5	Welded	5	Ethanol + distilled water
6	Seamless	5	Ethanol + distilled water
7	Welded	5	Moist argon
8	Seamless	5	Moist argon

After the specimens were removed from the sodium, they were cleaned by one of two methods:

1. Cleaning with ethanol until the hydrogen-producing reaction had virtually ceased, followed by ultrasonic cleaning in distilled water.

2. Bubbling argon through water at 50°C and then allowing the moisture-laden argon to impinge on the surface of the specimens. In moist-argon cleaning, the concentrated solution of sodium hydroxide that formed on the surface of the samples was allowed to remain there for 24 hr. The specimens were then ultrasonically cleaned in distilled water.

As a control experiment for the sodium exposures, sections of the Type 304L stainless steel welded and seamless tubing were given two different vacuum-ampule heat treatments: (1) sensitization at 650°C for 4 hr, followed by 5 weeks at 550°C; and (2) heat treatment at 550°C for 5 weeks without sensitization.





## B. Carburization Hypothesis

Standard chemical analyses and an electron microprobe made by Applied Research Laboratories were used to determine the carbon levels in the cladding of element C-249-16 and similar fuel elements.

Three sets of carbon-diffusion couples were prepared and exposed in separate tests to 550°C sodium (170°C cold trap) for 1 week to determine the potential for, and effects of, carbon mass transfer in liquid sodium at this temperature. The couples were prepared from ultrasonically cleaned 2-in. lengths of Type 304L stainless steel seamless and welded tubing. The 2-in. lengths were partially crimped shut at the bottom, and three steels of different carbon activity were placed inside them. Figure 4 shows a typical experimental tube-insert assembly.

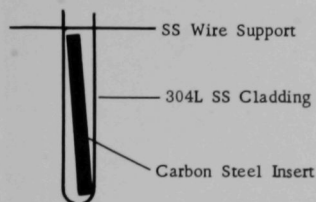


Fig. 4. Typical Assembly of Type 304L Stainless Steel Cladding and Carbon Steel Insert

In all, six couples were made, consisting of three different insert materials in three welded and three seamless tubes. The tubes and the inserts were weighed separately before assembly so that the extent of carbon mass transfer could be determined.

The inserts were (1) high-carbon ferritic steel (0.9-1.1 wt % carbon drill rod), (2) Type 304 stainless steel (0.06-0.08 wt % carbon), and (3) low-carbon ferritic steel (0.2-0.4 wt % carbon). The low- and high-carbon unalloyed steels essentially differed only in carbon concentration. Manganese, phosphorus, sulfur, and silicon were present in the two materials at nearly the same levels, so that the carbon concentrations give a fair indication of carbon activity in the steels. Table III lists typical analyses of the insert materials.

TABLE III. Analyses of Steels Used as Carbon Sources in Sodium-carburization Studies (wt %)

Element	Typical Low-C Steel	Typical High-C Steel Drill Rod
Carbon	0.20-0.40	0.90-1.1
Manganese	0.60-0.90	0.50-0.90
Phosphorus	0.040 max	0.040 max
Sulfur	0.050 max	0.050 max
Silicon	0.10-0.30	0.10-0.30

After the specimens were removed from the sodium, they were cleaned in ethanol and then cleaned ultrasonically in distilled water.



### III. DATA AND RESULTS

#### A. Cleaning Hypothesis

There were obvious differences in the microstructures of the as-received tube types (see Figs. 5 and 6). The grain size of the seamless tube was larger than that of the welded tube, and the inclusions in the seamless tube were more pronounced. The specimens heated in vacuum showed essentially the same metallographic features as those in specimens exposed to high-temperature sodium for the same period of time, no matter which sodium-removal technique had been used.

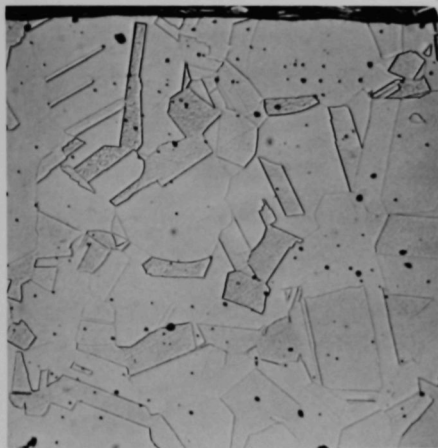


Fig. 5. As-received Type 304L Stainless Steel Seamless Cladding

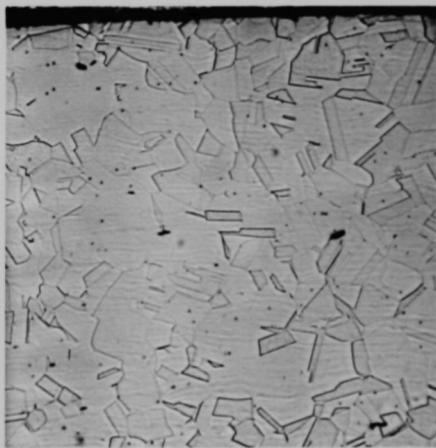


Fig. 6. As-received Type 304L Stainless Steel Welded Cladding

As shown in Figs. 7-10, the grain boundaries at the outer surface appeared somewhat wider and darker in all samples after sodium exposure, cleaning, and electrolytic etching. The heavier grain boundaries were found continuously around the outer circumference of the welded and seamless tubes. Later tests conducted by R. V. Strain showed that this behavior is characteristic of the Mark-I jacket tubing and that it is due to higher than average carbon concentration at the surface of the as-received tubing.

The microstructures of vacuum heat-treated seamless and welded Type 304L stainless steel cladding (Figs. 11 and 12) clearly reveal the carbon concentration gradient that is present in the as-received material.



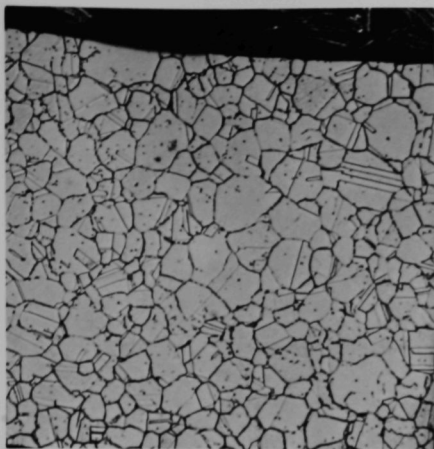


Fig. 7. Type 304L Stainless Steel Welded Tubing after Exposure to 550°C Sodium for Five Weeks Followed by Cleaning in Ethanol and Distilled Water

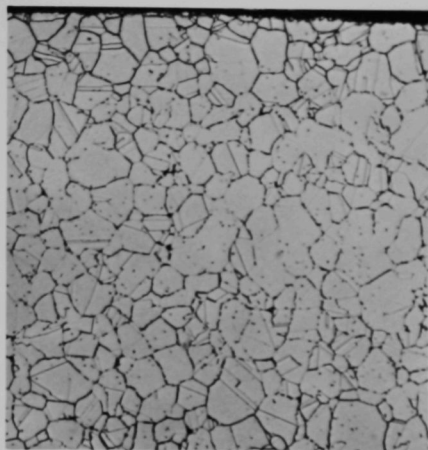


Fig. 8. Type 304L Stainless Steel Welded Tubing after Exposure to 550°C Sodium for Five Weeks Followed by Cleaning in Moist-argon Atmosphere

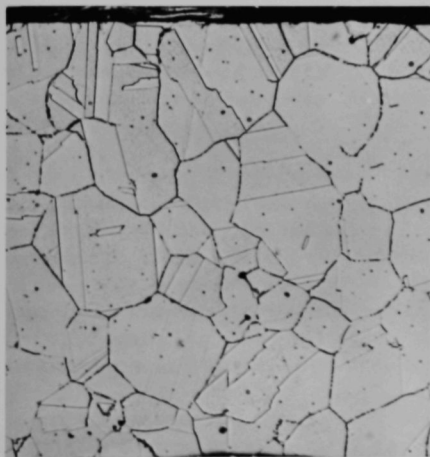


Fig. 9. Type 304L Stainless Steel Seamless Tubing after Exposure to 550°C Sodium for Five Weeks Followed by Cleaning in Moist-argon Atmosphere

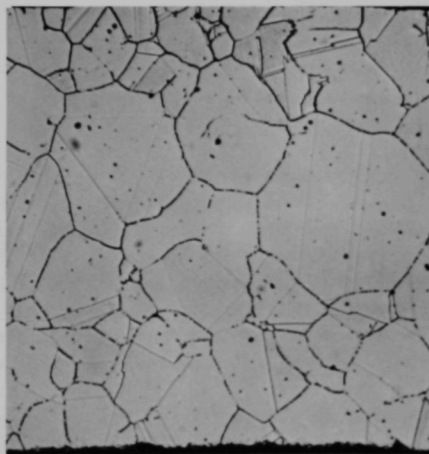


Fig. 10. Type 304L Stainless Steel Seamless Tubing after Exposure to 550°C Sodium for Five Weeks Followed by Cleaning in Ethanol and Distilled Water



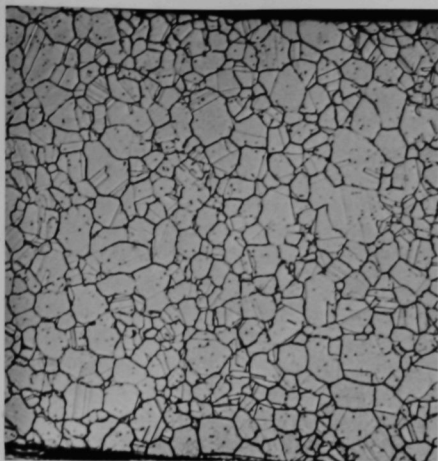


Fig. 11. Type 304L Stainless Steel Welded Tubing after Heat Treatment at 550°C for 38 days in Vacuum Ampule

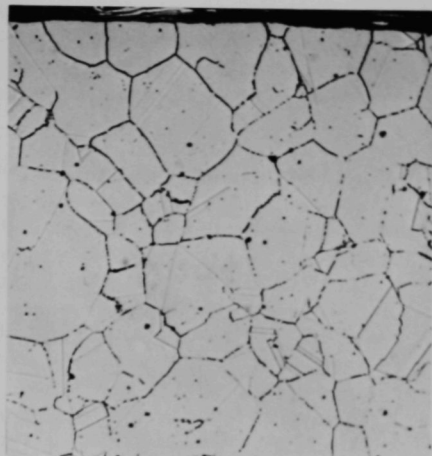


Fig. 12. Type 304L Stainless Steel Seamless Tubing after Heat Treatment at 550°C for 38 days in Vacuum Ampule

Some areas at the inner surfaces of the welded tubes showed somewhat enhanced grain-boundary etching with all cleaning techniques. In general, though, the enhanced etching attack at the inner surface of the welded tubes was much less evident than that at the outer surface. No accentuated grain-boundary etching was observed at the inner surface of the seamless tubes.

Two additional short tests were made of the ability of sodium hydroxide solution to attack intergranularly the cladding of element C-249-16. A polished and lightly etched section of this cladding was photographed and then exposed to 20 wt % NaOH solution at 100°C for 40 min. A second photograph of the same area revealed no change. A pellet of NaOH then was allowed to liquefy on the same polished section by moisture absorption. The concentrated caustic solution remained in contact with the metal for 18 hr. A final photograph of the same area again showed no attack.

This investigation into the cleaning hypothesis of cladding damage indicates that no normal variation in cleaning procedure could have resulted in the type of attack experienced by element C-249-16. The microstructures that develop when stainless steel is exposed to clean sodium or vacuum result from the time at temperature and are independent of the presence of the sodium and the subsequent sodium-removal technique.

#### B. Carburization Hypothesis

Bulk chemical analyses of element C-249-16 indicated an increase in carbon content from an initial value of 0.025 wt % to a final value of





0.035 wt %. Because the damage to the grain boundaries was concentrated in the outer 25% of the thickness of the metal, the increase in carbon concentration in this zone, if it were the causative element, would be substantially greater than the increase in the carbon concentration in the bulk of the metal.

Several samples of element C-249-16 were examined in the electron microprobe. From this examination, it was concluded on the basis of point counts that (1) the carbon concentration was higher in the grain boundaries than in the matrix, (2) the iron concentration was lower, and (3) the concentrations of chromium, nickel, manganese, silicon, nitrogen, and oxygen were the same in the grain boundaries as in the adjacent matrix. A similar examination of reactor-exposed pieces of Type 304 stainless steel from a control-rod thimble showed no detectable increase in carbon concentration at the grain boundaries, even though the sample had become sensitized. The inference was clear that the cladding of element C-249-16 had been selectively carburized at the grain boundaries, although other stainless steel exposed to the primary sodium in EBR-II had not.

Ex-reactor couples of carbon steel and Type 304L stainless steel were prepared and exposed to determine if the microstructural features of the cracked element C-249-16 could be duplicated. Table IV summarizes the gravimetric changes resulting from these experiments.

TABLE IV. Results of Experiments with Carbon-diffusion Couples

Tube Type (all 304L SS)	Insert Material	Change in Tube Weight, mg/in.	Change in Insert Weight, mg/in.	Calculated Carbon Content of Tube after Test, avg %	Analyzed Carbon Content of Tube after Test, avg %
Welded	Drill rod	+1.14	-1.35	0.22	-
Seamless	Drill rod	+1.12	-1.41	0.22	-
Welded	Low-carbon rod	+0.41	-0.82	0.11	-
Seamless	Low-carbon rod	+0.36	-0.82	0.08	-
Welded	304-SS control	-0.08	-0.08	-	-
Seamless	304-SS control	0	-0.09	-	-
Welded	Drill rod	+0.22	-0.48	0.073	0.083
Welded	None	0	-	0.03	0.024
Welded	Drill rod	+0.30	-0.55	0.088	0.083
Welded	None	0	-	0.030	0.023
Welded	Drill rod	+0.40	-	0.112	0.089
Welded	None	+0.03	-	0.039	0.024

Notes: Oxygen concentration in sodium as  $\text{Na}_2\text{O}$  was 2-7 ppm (by distillation).  
Couples were exposed separately for one week at 550°C.

Mass transfer of carbon definitely occurred from the carbon steel inserts to the seamless and welded tubes. When the insert was a drill rod, the cladding was carburized selectively along the grain boundaries of the inner surface and to a depth from one-third to one-half of the wall thickness (see Figs. 13 and 14). The grain boundaries were heavily delineated by continuous precipitates in both welded and seamless specimens. Even in



the as-polished condition, precipitates were very evident, along with an occasional boundary separation similar to that observed in the cladding of element C-249-16 (see Fig. 15).

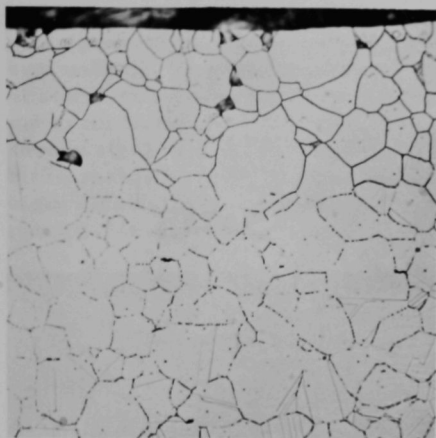


Fig. 13. Type 304L Stainless Steel Welded Tubing after Exposure to 550°C Sodium for One Week with Carbon-drill-rod Insert

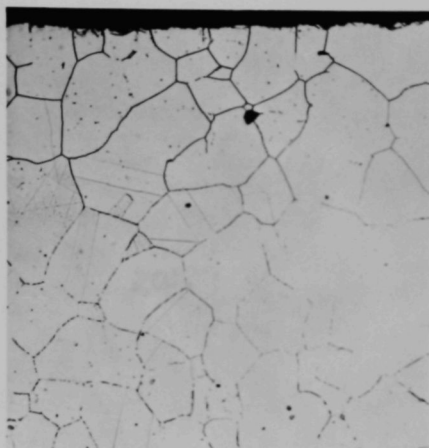


Fig. 14. Type 304L Stainless Steel Seamless Tubing after Exposure to 550°C Sodium for One Week with Carbon-drill-rod Insert

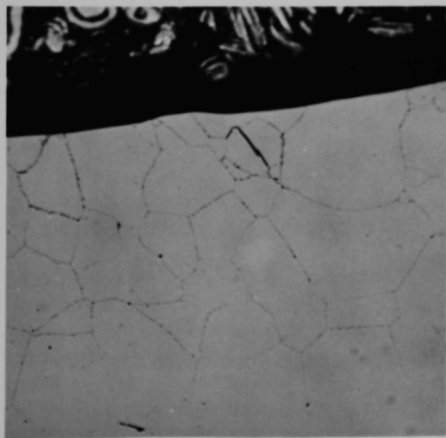


Fig. 15. As-polished Type 304L Stainless Steel Welded Tubing after Exposure to 550°C Sodium for One Week with Carbon-drill-rod Insert. (Reduced photographically by 10%)

However, the widening of the grain boundary upon etching was not nearly as severe as the heavy ditching observed in the cladding of element C-249-16, as shown in Fig. 1. That cladding showed very wide grain boundaries at the surface, but only to a depth from about one-fourth to one-third of the wall thickness. Also, the attack by the etching agent stopped more abruptly in the C-249-16 cladding than in the experimental specimens. However, the similarity in microstructure between the test and in-reactor specimens was evident.

By gravimetric calculations, it was determined that the amount of carbon transferred to the Type 304L stainless steel in the experimental tests had increased the bulk carbon



level of the steel to more than 0.2 wt % carbon (assuming all weight change to correspond to carbon transfer). This increase was greater than that experienced in the reactor.

A low-carbon steel rod was used as an insert material in an effort to more closely simulate the damaged tube structure. The resulting microstructures of the etched specimens (Figs. 16 and 17) revealed the same general widening of grain boundaries that was apparent in the microstructures of stainless steel that had been coupled with the drill-rod inserts. However, the grain boundaries near the surface were more heavily attacked when low-carbon steel rods were used, and the depth of the attacked zone was less. In these respects, the microstructures obtained using low-carbon steel inserts more closely resembled the microstructure of the cladding of element C-249-16. This can be seen by comparing Figs. 1 and 16.

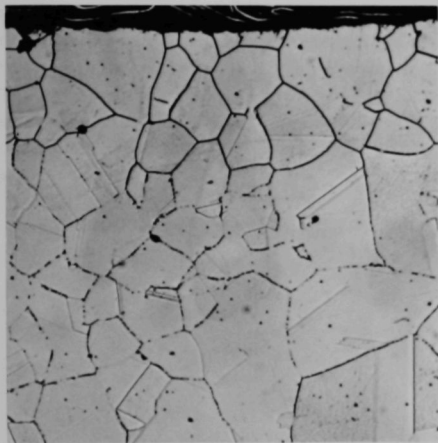


Fig. 16. Type 304L Stainless Steel Seamless Tubing after Exposure to 550°C Sodium for One Week with Low-carbon Steel Insert

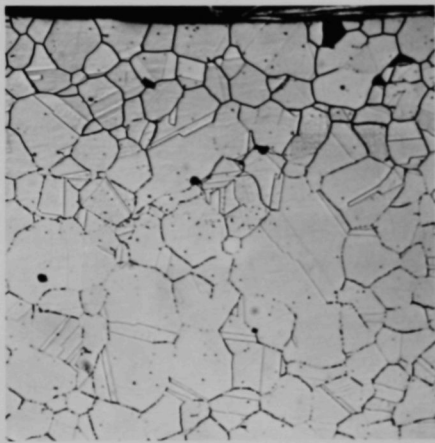


Fig. 17. Type 304L Stainless Steel Welded Tubing after Exposure to 550°C Sodium for One Week with Low-carbon Steel Insert

The specimens of Type 304L stainless steel tubing that contained Type 304 stainless steel inserts showed none of the grain-boundary attack observed with the drill-rod inserts. This observation suggests that the potential for carbon transfer between these two metals is small. This behavior is consistent with the view that the carbon activity in both metals is essentially the same. Differing bulk carbon concentrations vary only the equilibrium amount of the second phase, not the carbon activity.

The observation that the in-reactor attack appeared only in isolated areas in the fuel cladding suggested that a carbon source was close to the



areas of attack. A carbon source further away from the cladding (e.g., a high-carbon material elsewhere in the sodium system) would cause continuous carburization around the circumference of the cladding.

To evaluate the postulate that the carbon source was near the cladding, Type 304L stainless steel seamless and welded tubes were lightly streaked with either Apiezon-N grease (100  $\mu\text{g}/\text{in.}$ ) or a film of dry graphite applied by spraying with a commercial graphite aerosol preparation. After this contamination, the samples were exposed to 550°C sodium in the circulating autoclave for one week.

Subsequent microexamination revealed the same selective carburization as found in the experimental diffusion couples. However, the attack was restricted to the area near the site of contamination. The carburized area was larger than the contaminated region, because of physical smearing and surface-transport mechanisms in the sodium. A distinct break between the carburized and uncarburized regions of each tube was evident. A 45-sec etch in 5% oxalic acid revealed a striking similarity in microstructure between the seamless tube contaminated with Apiezon-N and the cladding of element C-249-16. Figures 18-22 are photomicrographs of the specimens contaminated with the two carbonaceous substances; these should be compared with Fig. 1.

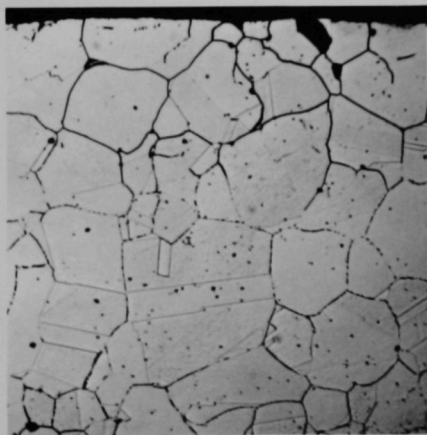


Fig. 18. Type 304L Stainless Steel Seamless Tubing That Had Been Streaked with Apiezon-N Grease and Exposed to 550°C Sodium for One Week (15-sec etch in 5% oxalic acid)

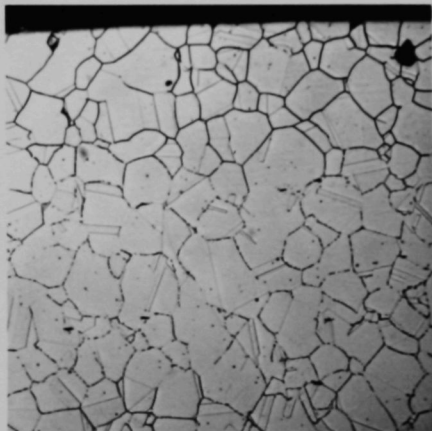


Fig. 19. Type 304L Stainless Steel Welded Tubing That Had Been Streaked with Apiezon-N Grease and Exposed to 550°C Sodium for One Week





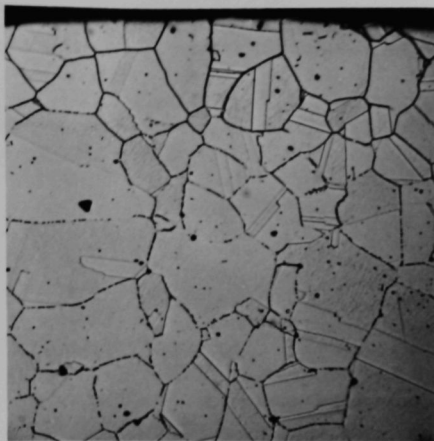


Fig. 20. Type 304L Stainless Steel Seamless Tubing That Had Been Streaked with Dry Graphite Film and Exposed to 550°C Sodium for One Week

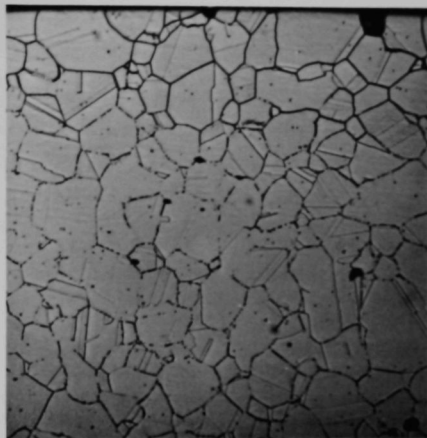


Fig. 21. Type 304L Stainless Steel Welded Tubing That Had Been Streaked with Dry Graphite Film and Exposed to 550°C Sodium for One Week

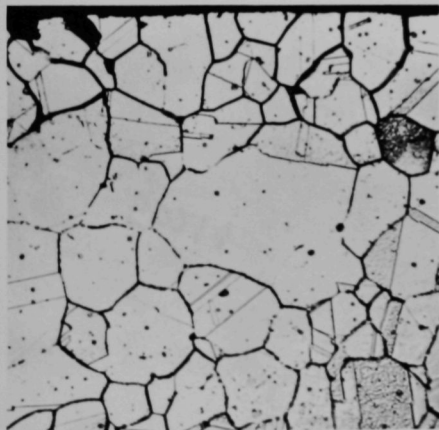


Fig. 22

Type 304L Stainless Steel Seamless Tubing That Had Been Streaked with Apiezon-N Grease and Exposed to 550°C Sodium for One Week (45-sec etch in 5% oxalic acid)

#### IV. CONCLUSIONS

Neither cleaning nor storage of the fuel element contributed significantly to the selective grain-boundary attack observed at the cladding surface.

The carburization explanation for the attack noted in the cladding of element C-249-16 appears to closely fit the observed facts:



1. Chemical and microprobe analyses revealed that a carbon-concentration increase was associated with the areas of attack.
2. Contamination of the stainless surface with a carbonaceous substance before high-temperature sodium exposure produced a microstructure similar to that observed near the attacked areas.
3. The isolated occurrence of the attack suggests localized surface contamination, very possibly associated with the manufacturing of the tubing. However, there is no way to distinguish between grease left on the tube during manufacturing heat treatments and accidental contamination of the completed fuel element before it was inserted into its subassembly.

#### ACKNOWLEDGMENT

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